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# Liquid-Metal-Bonded Gap for Light Water Reactor Fuel Rods

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# Abstract

In this research, liquid metal composed of 33 weight percent each of Pb, Sn, and Bi is explored as an alternate pellet-cladding gap filler to conventional helium. Since the thermal conductivity of liquid metal (LM) is ~ 100 times that of He, calculations from the HEATING 7.3 heat transfer code showed that the pellet centerline temperature is reduced by as much as 400 $^{o}$C for a rod linear power of 270 W/cm. Fission gas release is also reduced due to lower fuel temperatures. In addition, pellet-cladding interaction can be eliminated by creating a large initial gap by appropriately adjusting the pellet diameter. Results from the fuel behavior computer code FALCON indicated that in order to achieve a burnup of 6 atomic % and to avoid pellet-cladding contact, the required initial cold, radial, LM-bonded gap is ~ 150 microns.

In the experimental work during the first year of the project, methods have been developed to eliminate voids in the LM bond. The term "void" means an unfilled region in the gap formed due to surface tension and nonwetting effects of the LM with the fuel and cladding. Since voids are essentially vacuum, their presence in the gap may cause local fuel overheating due to severely hindered heat transfer. It was discovered that by applying pressure (up to ~ 100 psig) to the interior of the fuel rod after submerging pellets in a pool of LM before pushing them into the rod, voids can be collapsed for both types of pellets – dished-ended ones (PWR) and flat-ended ones (BWR). However, when the rod is only partially filled with LM before pellet loading (a method that provides the least disruption in manufacturing process), pressure can eliminate voids for the dished pellet case, but not for the flat-end pellets.

To non-destructively measure LM bond integrity, an eddy current testing technique was utilized. It was found that voids cannot be located by <u>axial</u> scans using ZETEC's pencil probe with a frequency of 100 Hz,. However, <u>circumferential</u> scans done at a constant elevation showed promising void-detection ability. Signals from pellet-pellet interfaces can also be precisely picked up by the pencil probe with an axial scan.

Future work includes: 1) Improving the eddy current test coil characteristics and frequency so that void signals can picked up when scanned axially. 2) Studying the effects of fission gas release on LM bond integrity using porous alumina pellets (a surrogate for $UO_2$ fuel) in a glass tube. 3) Fabricating a 50-cm long LM-bonded fuel rod

with radial gap size of ~ 100 microns of high LM bond integrity. 4) Performing a neutronics analysis to determine the uranium enrichment needed to offset the effect of reduced pellet diameter for the same nuclear performance. 5) Calculating the incubation time for fission product release and the release rate thereafter.

## 1. Introduction

In light water reactors, $UO_2$ pellets are stacked in Zircaloy cladding tubes with a predetermined space or gap between the fuel and the cladding. The purpose of the gap is to accommodate fuel swelling and clad creep down without introducing excessive heat transfer resistance. Wright et al first proposed the use of a liquid metal (LM) bond in the fuel-cladding gap of fuel rods of light-water reactors (1, 2). The alloy is composed of 33 weight percent each of lead, tin, and bismuth, chosen for its low melting point (~120$^{o}$C), its lack of chemical reactivity with $UO_2$ and water, and its high thermal conductivity ( ~100 times greater than that of He). One of the advantages of the liquid-metal bonded gap is reduction of the fuel temperature. This reduces fission gas release since the diffusion of fission gas is thermally activated. Finally, by appropriately adjusting the initial gap thickness, pellet-cladding mechanical interaction over the fuel lifetime can be completely avoided. However, the LM filler approach has some potential disadvantages. First, the complexity of fuel rod manufacturing process is greater due to handling LM and inserting it into fuel rods. Second, voids can develop in the gap during the manufacturing process due to surface tension and wetting effects with pellet and cladding surfaces. Since the voids are essentially high vacuum pockets, heat transfer from the fuel to the cladding becomes locally severely hindered, causing local fuel hot spots. Third, fission gas, once released into the gap, may have the similar consequences due to its very poor thermal conductivity.

In this year's work, approaches to completely filling the pellet-cladding gap with LM were explored, and methods of quality control in an actual fuel element fabrication process were also investigated.

## 2. Approach

Prior to testing Zircaloy cladding, clear pyrex glass "cladding" with a precisely known ID was used to visualize the formation and completeness of the LM bond. Two fabrication methods that provide the least disruption of the classical fuel fabrication procedures were developed: the *partial immersion* and *full immersion methods*. In the former, only the cladding tube is partially filled with liquid metal prior to insertion of the pellets. In the latter method, the cladding tube and the pressure fitting above it were filled with liquid metal. The partial immersion method uses less LM and avoids overflow of LM during pellet insertion, but is not as effective in removing entrapped air bubbles as is the full immersion method. However, this method is not as adaptable to commercial fuel manufacturing as the partial immersion method.

Overpressure applied to the inside of the cladding was used to eliminate voids in the gap during fabrication. Gaps of different sizes were tested to determine the effect of this parameter on bond integrity. For quality control, a non-destructive examination (NDE) technique to assess LM bond integrity when fuel pellets are clad with Zircaloy was tested.

Two computer-code calculations were performed. The first was to determine the initial gap size needed to prevent pellet-cladding mechanical interaction (PCMI) until 6% burnup; the second determined the temperature rise of the fuel due to voidage of the LM in the gap.

## 3. Progress this year

### 3.1 Experimental

The partial immersion method and the full immersion method are very similar. The only difference is the way pellets were loaded in. For both cases, a simulated fuel rod made of a clear pyrex glass tube with a precisely known ID was attached to a pressure fitting and sealed by a rubber O-ring (see Fig. 1). Photographs of the apparatus are shown in Figs 2a and 2b. A circular Teflon® plug ( ~ 1 cm height, milled to fit the tube ID) was used to close the bottom of an open-ended tube. For a closed-end tube as shown in Fig. 2a, the plug was not necessary. To prevent ejection of the glass tube from the pressure fitting, a lower end plate was used (see Fig. 2b). The unit was heated up by half of a clam-shell resistive heater to about 150-170 $^{o}$C (tube inside temperature). The temperature measurement was done by inserting a thermocouple to the inside of the tube.

For the partial immersion method, the LM was poured in to fill ~ ¼ of the height of the tube. Two types of pellets were used: BWR type (flat ends), and PWR type (dished ends). Pellets were pushed into the rod one by one. A total of 10 pellets of each type filled in the rod for each run. The stack height was 15 cm for PWR, and ~ 9 cm for BWR. The O-ring and cap were screwed into the pressure fitting, the upper fitting was attached, and connected to a gas cylinder. Varying pressures were applied the interior of the rod and the void sizes at different pressures were measured using a metal ruler (bent to a circular shape) that slid up and down the tube.

For the full immersion technique, the glass tube and pressure fitting (reservoir) were completely filled with LM. One pellet at a time was submerged in the LM in the reservoir and turned several times before being pushed into the glass tube. This ensured that there would be no air bubbles attached to the pellet surface. Void sizes at different pressure loadings were recorded.

Cladding of the type used in nuclear reactors provided by Siemen (Zr-2 standard throughwall type) was also tested. Two samples were sent to the NDE center in EPRI, North Carolina for eddy current testing. One was prepared by the partial immersion method using dished pellets and with an applied pressure of ~ 100 psig. This sample will be called the "no-void" sample thereafter. The other specimen was prepared exactly the same way as the first one except that no external pressure was applied. This sample will be called the "void" sample from now on. The eddy-current instrument used was ZETEC's MIZ-27. Two kinds of probes were used to acquire data: an encircling probe and a pencil probe. The samples were subsequently evaluated destructively at UC Berkeley. The Zry cladding was milled ~ 180$^{o}$ apart and was separated, exposing the LM coating and the pellets.

The Bi-Pb-Sn alloy with 33 w/o each was chosen for this application because the composition represents the eutectic point which has the lowest melting point (~120$^{o}$C). Studies done by Dubecky also revealed that the compound is compatible with $UO_2$ and Zircaloy for the range of the operating temperature (2). The compound also shows low neutron absorption cross section, which is crucial for the neutron economy.

## 3.2 Computation

ANATECH company ran the FALCON fuel behavior code in order to determine the initial gap size to avoid PCMI at 6 % atom burnup with the LM bond. The LM bond was simulated by changing the thermal conductivity of He gas to 100 times its nominal input value. The radial initial cold gap size at BOL to satisfy the above criterion was found to be 150 µm. By comparison, the usual hot gap thickness in LWRs is half of this value, and gap closure occurs at the end of the first cycle.

Fuel hot spots caused by voids of different sizes in LM bond were studied using the HEATING 7.3 heat transfer code (3). All the runs assumed a fuel pin with a continuous pellet column and with a constant cladding outer surface temperature of 295°C. Heat transfer by radiation across the void portion of the gap was ignored (because there were difficulties incorporating radiative heat transfer into HEATING 7.3). Fission energy deposition was assumed to be uniform. The linear heating rate was uniform axially at 270 /cm. A Westinghouse BWR Mk-1 fuel pin geometry, accounting for thermal expansion, was used for the code input. The initial hot radial gap size was ~ 100 microns. Oxides and surface roughness were ignored. Temperature distribution, fuel and cladding geometries, thermal and mechanical properties of fuel pin constituents were constant in time. The thermal conductivity of the pellet was assumed to be of the form:

$$k_{fuel} = \frac{1}{a + bT} \tag{1}$$

where a = 0.004 m-K/W, b = $2.25\times10^{-4}$ m/W[1], and K is in Kelvin.

One rectangular void with a height of 1 cm and variable fraction of the pellet periphery is placed in the liquid metal region. The pellet, LM, void, and cladding zones were discretized as required by the program, and the code was run under a steady-state condition. Results are shown in the following section.

[1] These values are obtained from major nuclear component manufacturers. The figures used here represent the average.

## 4. Results

Two Zircaloy-clad specimens were sent to EPRI's NDE center for analysis by the eddy current method. The specimen fabricated under 100 psig pressure (the "no void" specimen) and the specimen fabricated with no overpressure (the "void" specimen) both had nominal LM bond thicknesses of 94 μm. Seven pellets were in the void sample and ten pellets for the no-void sample. The pellet stack height was 15 cm for the no-void specimen, and 10.5 cm for the other.

### 4.1 Partial immersion with dished pellets

Figures 3a – 3g show the results of destructive analysis of the two samples returned from EPRI following eddy current testing.

The three photos in Figs. 3a – 3c cover the entire length of the "no-void" LM-bonded pellet stack. The pellet stack is shown at top with an intact (frozen) LM bond. The Zircaloy tubing removed from the pellets is shown below. It is deformed in the process of removing it from the pellet column. Very thin cracks in the LM bond are seen at the pellet-pellet interfaces. In these specimens, any voids were completely eliminated by pressurization, as it was observed in the glass runs. The bonding was also complete on the opposite side not shown here. The irregularities in the LM bond along the circumference were probably caused as the pellets were forced out of the cladding. The patterns did not appear when pellets were not disturbed. Figure 3d reveals liquid metal in the dished portion between pellets. It was observed that LM even went into the "40" carving on the pellet.

For the "void" specimen, several voids were observed and the results are depicted in Fig 3e – 3g. These results qualitatively agree with the glass-cladding tests in the substantial uncovered area in the gap. Note that some portions of the pellets were absent from their sites in the LM bond. They fell off while the cladding was pulled apart.

With the partial immersion loading technique using dished pellets, air was trapped on the pellet surfaces when the pellets were pushed into the LM partially filling the cladding tube. It is believed that these air bubbles moved into the dished portion between pellets during pressurization. That was why they disappeared from the LM bond. The cylinder with the markings adjacent to the pellet stack in Figs. 3f and 3g is a plastic straw with markings intended to show how the eddy current response corresponded to irregularities in the fuel rod. The dots represent pellet-pellet interfaces and the squares represent NDE signals that possibly correspond to the voids observed in the destructive analysis.

**4.2 Partial immersion with flat pellets**

For the pellets with flat ends prepared by the partial immersion method, voids were observed even with pressurization up to ~ 100 psig, with a 190 μm radial gap thickness. It is believed that air bubbles stuck to the pellet surfaces had no place to go. Therefore, they remained and gave the same appearance as true voids. Higher pressures and a more vigorous pellet insertion method will be tried in the future to see if these air bubbles can be eliminated

**4.3 Full immersion with dished pellets**

The sequence of eight photographs in Figs 4a – 4h show a test in glass cladding, in order of increasing overpressure. The full immersion method was used with the dished type of pellets. The radial gap size was ~ 100 microns. It was observed that high pressure could collapse the voids. At initial atmospheric loading, the void height was usually ~ 1 pellet height (not seen in the pictures, but was observed in several other runs). The voids were roughly rectangular-shaped or wedge-shaped and extended from 0% to ~ 75% of the circumference. Since there was no air trapped inside the voids with the full immersion method, the voids were eliminated as increasing pressure overcame the surface tension. The surface tension of the liquid metal combined with its inability to wet either $UO_2$ or the native oxide coat on the Zircaloy inner surface are responsible for the creation of voids.

Figure 5 shows the plot of void fraction vs. pressure for three different radial gap sizes, 100, 145, and 200 μm. Void fraction is defined as the fraction of the circumferential area in the pellet regions that voids occupy (all voids are about the height of one pellet). The full immersion method was used with dished pellets loaded in a glass tube. The following trends were observed: Void fraction decreases rapidly with the initial increase in pressure. In most of the runs, the fraction becomes less than 0.5% at pressure of 70 psig. Further, a smaller gap size yielded a bigger void fraction at the beginning. At higher pressures, the curves virtually merged together, irrespective of the gap width. The pressure effect was reversible: the voids reappeared as the pressure was reduced.

**4.4 Full immersion with flat pellets**

Flat pellets were also loaded using full-immersion with 190 microns gap size. It was observed that voids were collapsed with overpressurization. Since voids had no air inside, pressure could overcome the surface tension, as in the case with dished pellets.

**4.5 Effect of Voids on Hot Spots in the Fuel**

The results from the HEATING 7.3 code are presented here. Figure 6 shows the plot of the maximum fuel temperature as a function of void fraction (i.e., the fraction of the pellet circumference not covered by LM). The maximum fuel temperature no longer occurs at the fuel center, but moves towards the voided region. For a void fraction greater than 40% (assuming 1-cm height and a rectangular shape), the maximum temperature exceeds that of He case. Therefore, the thermal benefit of the LM bond is lost (at least locally) if a void displaces more than 40% of the surface. However, if the LM bond is circumferentially and axially intact, the reduction in the maximum fuel temperature is ~400$^{o}$C (for a linear heat rating of 270 W/cm).

**4.6 Results of NDE testing**

After the cladding was separated from the frozen LM and the pellet stack, signals from the eddy current testing were matched with what was observed. The frequency used to feed the probe was 100 kHz because it gave the best signal.

Figure 7a shows the signal from the “no-void” specimen. The crescent-shaped figure in the gridded area indicates a region of reduced electrical conductivity at an axial elevation between the two dotted horizontal lines on the left. Figure 7b matches the vertical output shown on the leftmost column in Figs 7a and 7b. It was observed that the encircling probe could very effectively differentiate between the base metal (i.e., the empty Zircaloy tube) and region completely filled with LM but containing no pellets since it produced a sharp rise at the interface between the two regions. However, in the LM-filled gap region, the reading was not that clear. The small bumps in the pellet-filled zone indicated some irregularities in the conductivity of the cross-section.

For conventional eddy current applications, professionals are more interested in the shape of the signal in the impedance plane display (the banana-like contour in the big square one on the right of Fig. 7a) because it infers characteristics of cracks in different materials. Since we are only looking for changes in the electrical conductivity, the signal in the leftmost column suffices for our application.

In order to obtain a better signal in the pellet region, a pencil probe was used. The probe, which looked like a pen, was slid on the specimen either axially or circumferentially. Thus, it can detect local irregularities better than the encircling probe, which only sees the average of the cross-section. The signal is shown in Figure 7c. By destructively examining the specimen (Fig 3a – 3d), the bumps on the left hand vertical traces corresponded perfectly to pellet-pellet interfaces. The sharp drop in the vertical scan on the second column is not of our interest. It is a signal from another channel with different frequency. We did not use it in this analysis. A circumferential scan on the specimen surface was performed and a bump in the signal at one elevation (Fig 7d) corresponded to a pellet that was substantially off center.

The eddy current signals from the “void” specimen are presented in Figs. 8a – 8f. Fig 8a shows the signal from the encircling probe. With matching presented in Figure 8b, it is seen that the encircling probe did a good job for this specimen in pinpointing the interface location between the empty tube and the portion filled with LM. However, it still gave poor readings in the pellet area.

For a greater detection capability, the pencil probe was used to scan the specimen axially for four locations spaced ~ $90^{o}$ apart. Figures 8c – 8f show the signals. By comparing the signal with the cut-open specimen, signal peaks corresponded perfectly to pellet-pellet interfaces. These peaks indicate a higher presence of LM locally, probably from the LM that entered the dished portions of the pellets. It can further be inferred that LM must have entered the cracks in the pellets proper, because some peaks from the pencil probe corresponded to visually observed cracks.

In Fig. 8c – 8f, changes in the signal when the probe passed over gap voids however, were inconclusive. Sometimes the signal went up when passing over a void, but sometimes it went down or remained relatively unchanged. The correlation between the eddy current signal and the appearance of the cut-open specimen was not strong enough to draw any conclusion. Ideally, it would be expected to see the signal drop when passing through a void since it has poor conductivity compared to LM. The reason why it was not observed is most likely that the coating of LM around the pellet is so thin that the minute increase in local conductivity cannot be picked up. To compare, the cladding thickness is ~ 1500 microns, and the maximum gap thickness is 188 microns (assuming the pellet touches the wall on the opposite side), so the cladding is ~ 8 times thicker than the LM layer. For a perfectly centered pellet, the LM bond is only 1/16 of the cladding thickness.

The pellet-filled portion of the specimen was also scanned circumferentially at six elevations with the pencil probe. Figure 8g – 8l shows the signal responses. By matching the signals with the specimen, it was concluded that the two peaks at each elevation corresponded roughly to the beginning and the end of a void, the valley between the two being in the void region. (The pairs of dotted horizontal lines did not mean anything for this analysis. We moved them there because we wanted to look at the portion of the signal in the impedance plane display.) Therefore, a pencil probe scanned circumferentially seems to be able to locate voids. However, the signal still does not agree with our expectation because of the following reasons: First, it was concluded in the above paragraph that the axial scan cannot reveal the presence of voids presumably because the LM layer is too thin. Second, if the circumferential scan were, for any reason, able to differentiate signals from the thin layer of LM from voids, we would expect to observe signals dropping down between the two peaks (due to no conductivity in voids), and going up outside (due to conductivity of LM coating).

The circumferential scan technique appears to be superior to the axial scan method in responding to voids in the LM bond. However, since eddy current inspection is an inference based test, appropriate reference standards are required for actual test conditions. A plan to produce reference standards to match the signal is discussed in the next section.

## 5. Future work

1. Since in the actual fuel rod manufacturing process, it would be convenient to move the probe from the top to the bottom of a fuel rod and analyze the response, the axial scan using a pencil probe will need further study. Consequently, more runs with glass cladding will be made so that voids are observed when passing the probe above them. This will provide a better match of the probe signal with voids than could be achieved by destructive examination of Zircaloy-clad pellets. The glass will have the same thickness as the Zircaloy cladding, and the gap size will be ~ 100 microns. We are planning to send some samples to ZETEC company where an engineer will help determine the best coil characteristics and settings on the instrument to achieve the best void detection capability. Hopefully, this will lead to a reliable method of detecting voids in the LM bond of Zircaloy-clad fuel rods.
2. Porous alumina pellets, substituted for $UO_2$ pellets, will be used to simulate and study the effect of fission gas release on the integrity of the LM bond in the fuel-cladding gap. The rough procedure will be: 1) load the pellets in a glass tube containing LM 2) heat up the tube to an appropriate temperature to "release" a desired amount of "fission gas." Actually, the air contained in the open porosity of the alumina pellets will be driven out simply by the ideal-gas law requirement that less gas must be contained in the pellet pores at the higher temperature.
3. We will finalize the fabrication method and will demonstrate fabrication of ~50 cm high fuel rod with LM bond of high integrity. The gap size will be ~ 100 μm. We will perform eddy current testing to confirm LM bond integrity.

4. We will perform a neutronics analysis to determine the effect of a larger initial gap (compared to the conventional He-filled rod) and the resulting smaller fuel pellet diameter on the U-235 enrichment needed for the same nuclear performance.
5. We will calculate the effect of eliminating the gap thermal resistance, and the consequent reduction in fuel temperature, on the incubation time for fission product release and the release rate thereafter.

## 6. Conclusions

1. The full immersion method with overpressurization ( ~ 70 psig and above) can eliminate voids for both flat and dished pellets.
2. Partial immersion with overpressurization can eliminate voids for the dished pellet type, but so far cannot for the flat ones (higher pressures will be tried).
3. Calculations done by ANATECH show that, with LM bonding, the gap size required to achieve to 6% atomic burnup without PCMI is ~ 150 microns.
4. Results from HEATING 7.3 indicate that a void fraction of more than 40% for a 1-cm height void will result in the pellet maximum temperature exceeding that in the conventional He design. An intact LM bond reduces the centerline fuel temperature by ~400$^{o}$C for an LHR of 270 W/cm.
5. Eddy current testing with an encircling probe shows good detection ability to pinpoint the interfaces between the empty cladding, LM-filled cladding, and pellet-filled regions. However, it was not successful in detecting voids in the LM bond.
6. Eddy current testing with a pencil probe (axial scan) has a good detection capability to pinpoint the pellet-pellet interfaces.
7. Eddy current testing with an axial scan using a pencil probe does not have a good detection ability to locate voids.
8. Eddy current testing with a circumferential scan using a pencil probe provides a good detection ability to locate voids. However, this method may not be convenient for large-scale manufacturing processes.

# 7. References

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